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Abstract title: Pulsed Laser Deposition (PLD) of the Solar Cell Materials CZTS and CTS

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Abstract body: Abstract Body
CZTS (Cu_2ZnSnS_4) is a promising material for solar cell absorbers and consists of abundant and environmentally friendly elements. The efficiency of a cell based on this material has increased from 2% in 2001 to 8.8 % in 2015 [1]. CTS (Cu_2SnS_3) is a similar material, which recently has reached an efficiency of 4.3 % [2].

Pulsed laser deposition (PLD) is considered as a technique, by which a material with a complicated stoichiometry can be transferred from a target to a growing film in a vacuum chamber. It is a versatile technique, particularly suited to mixtures of elements, e.g. metal oxides and alloys. However, if a volatile component in the target material, e.g. oxygen, is lost by evaporation from the target or the film during the laser irradiation, the stoichiometry can be controlled by performing the deposition in a background gas, e.g. also oxygen.

However, for sulfides the deposition needs to be carried out in a background gas of H_2S which is explosive and toxic. Alternatively, the sulfurization can be performed by a post-(deposition) annealing treatment in sulfur atmosphere. This treatment is time-consuming and involves a careful optimization procedure.

We investigate a new approach to prevent the loss of sulfur in CZTS, which is by using a reactive sulfur beam during PLD of CZTS in vacuum. The sulfur cracker source provides a beam with a high concentration of reactive sulfur dimers (S_2) and sulfur trimers (S_3) during the growth by cracking long rings and non-reactive chains of S_8 molecules. We will show the first preliminary results from a simultaneous operation with a PLD set-up and a sulfur cracker in a vacuum chamber.

The physics behind the PLD process of chalcogenide materials CZTS and CTS is partly determined by the composition of the target materials (which are typically made sintered powders of binary compounds). Our results show that the deposition process is a combination of evaporation of the most volatile phases/elements and ablation of the most stable ones. In our case at low laser fluence ($F< 0.5J/cm^2$) the dominant process is laser-induced evaporation: films are very smooth and no (or very little) copper is transferred from the target. Indeed, the most volatile elements in the target are S and Zn (and SnS as a compound). On the other hand, at high fluence ($F> 1.5J/cm^2$) films are characterized by an over-stoichiometric Cu-content (with respect to the target) and severe sulfur loss. The region of stoichiometric transfer is found between these two values of the laser fluence. We have obtained an efficiency of the CZTS cells exceeding 2 % with the most appropriate film composition and expect to reach a higher efficiency with the sulfur cracker.

[1]S. Tajima et al., *Appl. Phys. Express* 2015, **8**, 082302

[2] A. Kanai et al. Jap. J. Appl. Phys. **54**, 08KC06 (2015)

[3] L.-C. Chen, in: Pulsed laser deposition of thin films, Wiley (1994) p. 167.